

Journal of the Air & Waste Management Association



ISSN: (Print) (Online) Journal homepage: https://www.tandfonline.com/loi/uawm20

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To cite this article: Chelsea Berg, Brian Crone, Brian Gullett, Mark Higuchi, Max J. Krause, Paul M. Lemieux, Todd Martin, Erin P. Shields, Ed Struble, Eben Thoma & Andrew Whitehill (2022): Developing innovative treatment technologies for PFAS-containing wastes, Journal of the Air & Waste Management Association, DOI: 10.1080/10962247.2021.2000903

To link to this article: https://doi.org/10.1080/10962247.2021.2000903





OPEN ACCESS Check for updates **REVIEW PAPER**



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Developing innovative treatment technologies for PFAS-containing wastes

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ABSTRACT

The release of persistent per- and polyfluoroalkyl substances (PFAS) into the environment is a major concern for the United States Environmental Protection Agency (U.S. EPA). To complement its ongoing research efforts addressing PFAS contamination, the U.S. EPA's Office of Research and Development (ORD) commissioned the PFAS Innovative Treatment Team (PITT) to provide new perspectives on treatment and disposal of high priority PFAS-containing wastes. During its sixmonth tenure, the team was charged with identifying and developing promising solutions to destroy PFAS. The PITT examined emerging technologies for PFAS waste treatment and selected four technologies for further investigation. These technologies included mechanochemical treatment, electrochemical oxidation, gasification and pyrolysis, and supercritical water oxidation. This paper highlights these four technologies and discusses their prospects and the development needed before potentially becoming available solutions to address PFAS-contaminated waste.

Implications: This paper examines four novel, non-combustion technologies or applications for the treatment of persistent per- and polyfluoroalkyl substances (PFAS) wastes. These technologies are introduced to the reader along with their current state of development and areas for further development. This information will be useful for developers, policy makers, and facility managers that are facing increasing issues with disposal of PFAS wastes.

PAPER HISTORY

Received July 23, 2021 Revised October 5, 2021 Accepted October 22, 2021

Innovative team approach

The presence of persistent per- and polyfluoroalkyl substances (PFAS) in the environment is one of the most pressing environmental issues of the 21st century (Lim 2019; Lindstrom, Strynar, and Libelo 2011; Pan et al. 2017; Sunderland et al. 2019). There is high interest in understanding and addressing PFAS contamination among industry, state and federal government, and internationally. In April of 2020, the United States Environmental Protection Agency (U.S. EPA) formed the PFAS Innovative Treatment Team (PITT) to explore innovative tools and methods for destroying all the carbon fluorine (C-F) bonds in PFAS-containing waste. This interdisciplinary team consisted of eleven scientists and engineers from the Office of Research and Development (ORD) and operated under the framework of a "Tiger Team" as used by The National Aeronautics and Space Administration (NASA) Dempsey et al. (1964) or "Skunk Works" teams as used by corporations Lockheed (2021). During the team's six-month lifetime, it investigated numerous technologies to address PFAS contamination in various waste streams, with a goal of exploring several promising and understudied technologies. The team was provided

with financial resources to initiate the study of different treatment technologies at the laboratory, pilot, or field scales. The work resulted in improved understanding and advancement of four innovative technologies to supplement ongoing EPA research into PFAS treatment technologies. These technologies are electrochemical oxidation (EO), supercritical water oxidation (SCWO), mechanochemical degradation (MCD), and gasification and pyrolysis. Efforts were also initiated to quantify PFAS destruction in traditional waste treatment processes such as conventional thermal incineration. Results from laboratory and field work initiated by or supported by the PITT are ongoing and will be presented in future publications.

In this paper, we provide an overview of PFAS waste disposal and destruction issues, a summary of the five PFAS-containing waste streams that were considered, a discussion of the four innovative technologies studied, and preliminary thoughts about the applicability of technologies to different waste streams. Technical results from laboratory and field tests of PFAS treatment by individual technologies will follow in future publications.

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The PFAS challenge

The nature of the C-F bond makes PFAS difficult to destroy (Lemal 2004; O'Hagan 2008; Tsang, Burgess, and Babushok 1998). The small size and high electronegativity of fluorine creates the strongest bond in organic chemistry when bonded to carbon, which results in fairly inert molecules with very low surface energies (Lemal 2004; O'Hagan 2008). Molecules and polymers containing many C-F bonds are generally highly stable and unreactive. C-F bonds are very rare in nature Goldman (1969), but are abundant in several classes of industrially-produced chemicals, including PFAS. The PFAS class of compounds includes thousands of different substances (Lindstrom, Strynar, and Libelo 2011) which differ primarily by the number of C atoms in the alkyl chain and the functional groups typically attached to the end of the chain.

The first reported synthesis of a C-F bond was in 1862 Borodin, (1862). In 1890, Moissan described perfluorinated alkanes (Moissan 1890a, 1890b), a new class of fluorinated molecules that had been synthesized and isolated. He noted that the fluorinated compounds were particularly inert and stable; tetrafluoromethane could only be destroyed completely by heating it over sodium (Moissan 1890a, 1890b). Tetrafluoromethane and fluorinated alkyl molecules were first isolated in 1926¹⁴, mass produced as precursors and refrigerants by the mid-1930's (Daudt 1935; Okazoe 2009), and incorporated in greases by 1940 Gaylor (1940).

Research, development, and application of PFAS accelerated at the start of World War II, with strong links to the Manhattan Project. Liquid perfluorinated molecules and inert and formable fluoropolymers played a vital role in uranium enrichment, protective coatings, and other military applications Okazoe (2009). Many of these compounds were first developed in the late 1930s (IG Farbenindustrie, 1937, Plunkett 1941). Following World War II, companies involved in the war effort patented and developed commercial uses for the growing number of PFAS. Over the last 70 years, PFAS have been used in a variety of products. Products making extensive use of PFAS molecules include the following: protective non-stick and stain-resistant coatings Dinglasan-Panlilio and Mabury (2006), surfactants, fire-fighting agents (DeYoung 1994; Roth et al. 2020), temperature resistant products, food-contact materials (Schaider et al. 2017; Trier, Granby, and Christensen 2011), cosmetics Schultes et al. (2018), and more (Lemal 2004; Prevedouros et al. 2006). These synthetic compounds are used by industry, consumers, governments, and militaries around the world.

The characteristics that make PFAS useful in a wide variety of products also make them extremely difficult to destroy. The C-F bond is resistant to breakdown from ultraviolet light, biological processes, and heat. As a result, PFAS have become widespread persistent pollutants found around the world (Cicerone 1979; Cordner et al. 2019; Giesy and Kannan 2001; Lindstrom, Strynar, and Libelo 2011; Martin et al. 2002; McCord and Strynar 2019; Ravishankara et al. 1993; Stock et al. 2004). Some PFAS are detrimental to the environment (Ghisi, Vamerali, and Manzetti 2019; Ivy et al. 2012; Pan et al. 2017) and human health (Sunderland et al. 2019; Szilagyi, Avula, and Fry 2020). People can be exposed to PFAS from contaminated drinking water, fish, and food packaging (Sunderland et al. 2019; Trier, Granby, and Christensen 2011). Dermal, organ, and inhalation exposure to PFAS are all possible, especially for workers involved in making or processing PFAS and PFAS-containing materials where inhalation is the most likely route (Heydebreck et al. 2016). The best studied perfluoroalkyl acids are perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA). These long-chain PFAS can bioaccumulate over time to reach harmful levels and are linked to liver, immunological, developmental, reproductive, endocrine, cardiovascular, and cancer effects (Council 2020). With PFAS being in the blood of 98% of Americans, (Calafat et al. 2007) and PFAS' associated adverse health effects (Eriksen et al. 2009; Grandjean et al. 2012; Lewis, Johns, and Meeker Strynar, and Libelo 2015; Lindstrom, Sunderland et al. 2019), it is important to find effective methods to reduce the amount of PFAS in the sources of our exposure.

PFAS are present in several high-volume waste streams. These include municipal solid waste, landfill leachate, wastewaters treated at wastewater treatment plants, sewage sludge and biosolids from sewage treatment, and both concentrated and diluted Aqueous Film Forming Foam (AFFF). Treating the PFAS in these wastes requires methods that ensure PFAS and other harmful chemicals do not continue to accumulate in people or the environment. It is relatively easy to remove the functional group from a PFAS molecule, and thus destroy the initial compound; however, this often results in the formation of a different PFAS or organofluorine compound (Krusic, Marchione, and Roe 2005; Krusic and Roe 2004). This can create misleading information about the ultimate destruction of PFAS. The destruction and removal efficiency (DRE) for a treatment method can be very high, but the C-F bonds may all still be intact Watanabe et al. (2018). C-F bonds can require temperatures over 1400°C to be



destroyed thermally (Tsang, Burgess, and Babushok 1998) or highly reducing or oxidizing conditions for destruction at lower temperatures (Costello and McCarthy 1984; Lebeau and Damiens 1926; Moissan 1890a), so PFAS and organofluorine byproducts can be released into the environment from most treatment methods. Many of these partially destroyed PFAS molecules can react and be oxidized in the atmosphere or groundwater to form more harmful species, such as carboxylic acid functionalized PFAS (Ellis et al. 2004; Martin et al. 2006; Prevedouros et al. 2006; Stock et al. 2004). Ideal destruction methods would mineralize all the fluorine in the PFAS, breaking all the C-F bonds, to ensure that no organofluorine byproducts are produced.

The disposal and destruction of PFAS in contaminated sources is a complex area of research. PFAS are found throughout the world in most every matrix, making one universal destruction technique nearly impossible. Each of the main sources of PFAS needs to be carefully examined to determine the best approach for remediation or destruction.

PFAS waste streams

To properly address the PFAS waste management issues, it was first necessary to identify significant PFAS-containing waste streams and to understand both the nature of those wastes and the magnitude of the PFAS issue regarding those wastes. The five waste streams the PITT focused on were (1) landfill leachate, (2) biosolids, (3) contaminated soils, (4) granular activated carbon (GAC) or ion exchange resins used to remove PFAS from other media, and (5) AFFF, both concentrated and diluted. Each of these waste streams will be summarized in this section to provide a context for the PFAS destruction techniques explored in the following section.

Landfill leachate

In 2018 the U.S. disposed over 310 million metric tons of waste in nonhazardous waste landfills (U.S. EPA 2020), of which at least half was municipal solid waste (MSW). MSW contains many consumer products known to contain PFAS, including food contact paper (lined to-go containers), textiles (carpets and clothing), cleaning agents, and non-stick cookware, among others. Since PFAS are not considered hazardous wastes and are not accounted for accordingly, other commercial or industrial wastes deposited in landfills may also contain these compounds.

Modern sanitary landfills are lined with a plastic membrane overlying low-permeability soil, such as clay, which limits groundwater contamination but requires treating the collected leachate. Because of the direct leaching of PFAS-containing wastes, landfill leachate tends to have elevated PFAS levels compared to municipal wastewater alone (Huset et al. 2011; Masoner et al. 2020). Lang et al. (2017) estimated 61.1 million m³ of leachate were generated across the U.S. in 2013, comprising between 563 and 638 kg of PFAS.

Landfill leachate also tends to have higher suspended solids, chemical oxygen demand (COD), ammonia, salinity, and alkalinity than municipal wastewaters (Kjeldsen et al. 2002). The high organic matter loading makes leachate difficult to treat on its own, and any treatment of landfill leachate specifically for PFAS may require pre-treatment for cocontaminants (Wiszniowski et al. 2006). Many wastewater treatment plants do not contain the necessary treatment technologies (specially designed granular activated carbon, ion-exchange resins, reverse osmosis, and nanofiltration systems) to fully remove PFAS from treated water (Aro et al. 2021; Pan, Liu, and Ying 2016). As a result, the PFAS compounds have the potential to remain in the treated effluent.

Biosolids

In addition to the treated wastewater, wastewater treatment plants must also safely manage the residual sludges produced during treatment processes. Influent wastewater streams generally contain PFAS in various concentrations, which are then detected in both the treated effluent water and in the biosolids and sludges (Aro et al. 2021; Chiavola et al. 2020; Dauchy et al. 2017; Gallen et al. 2018a; Loganathan et al. 2007a). In the U.S., solids and sludge are largely either incinerated (~16%), landfilled (~22%), or land applied (~52%) U.S. EPA (2019). For land application, time and temperature treatment of WWTP solids to form Class A or B biosolids is regulated by the EPA to reduce pathogens, vector attraction, and heavy metals before use as nutrients and soil conditioners (U.S. EPA 1994). Since 2001, a growing number of studies (Alder and van der Voet 2015; Chen et al. 2012; Gallen et al. 2018b; Loganathan et al. 2007b; Michigan Department of Environment, 2020; Navarro et al. 2016; Schultz et al. 2006; Sepulvado et al. 2011; Sinclair and Kannan 2006; Venkatesan and Halden 2013; Yoo et al. 2009; Yu et al. 2009) have shown PFAS in biosolids in the U.S. and internationally with typical results showing PFOA levels from <20 μg/kg to as much as 240 µg/kg dry wt. These surveys show a larger range in

PFOS levels generally from <30 μg/kg to over 1000 μg/ kg. The concentrations of individual PFAS in biosolids show a high spatial and temporal variability. As a result of several rare cases of land-applied biosolids containing high PFAS levels, some states in the U.S. (such as the state of Maine) limit the PFAS concentration of biosolids that are to be land applied. Addressing PFAS in biosolids in a way that allows beneficial reuse or resource recovery is a priority.

Contaminated soil

The heterogenous nature of soils creates complex interactions with PFAS. The soil's organic content and surface charges are the major contributors to PFAS retention. Soils with high organic content can interact with and retain PFAS through hydrophobic interactions, while ions in the soil can interact with PFAS electrostatically. The length of the PFAS chain is the main factor in the PFAS partitioning into soil. As the tail gets longer, the PFAS is retained more due to the increasing size and greater hydrophobicity. A charged functional group can also increase retention (Mejia-Avendaño et al. 2020; Xiao et al. 2019).

The strong chemical structure of PFAS and their bonding with soil makes them challenging to eliminate from soil environments. Washington et al. calculated global soil loadings for eight PFAS compounds (Washington et al. 2019). The combined estimated load for the eight PFAS compounds ranged from 1500 to 9000 metric tons, with mean estimates of approximately 1000 metric tons for both PFOA and PFOS. These results indicate that soil has the potential to be a reservoir for PFAS. Soil concentrations reported for PFAS-contaminated sites are generally orders-ofmagnitude greater than background levels (Brusseau, Anderson, and Guo 2020). Maximum reported PFOS concentrations for primary contaminated sites ranged upwards of several hundred mg/kg. For example, a maximum PFOS concentration of 373 mg/kg was measured at an AFFF source zone on a U.S. military installation (Brusseau, Anderson, and Guo 2020). PFAS soil concentrations for secondary-source contaminated sites (e.g., land application of municipal biosolids and use of contaminated ground water for irrigation) were about two orders of magnitude lower (Brusseau, Anderson, and Guo 2020).

Methods for PFAS removal from contaminated soils have been reviewed by Ross et al. (2018), Mahinroosta and Senevirathna (2020), and Bolan et al. (2021). Mature treatment technologies for soils include incineration, soil stabilization, excavation, and ex-situ thermal treatment Ross et al. (2018). Recent field tests focusing on sampling methods development have examined soil incineration of AFFF-contaminated soil as a secondary goal, cumulating in a proposed test method, OTM-45 (Merrill and Ryan 2021), and the Department of Defense's (DoD's) associated Strategic Environmental Research and Development Program (SERDP) final report for Project ER19-1408 to be released on SERDP's web site, www.SERDP.org. More experimental methods include soil washing, advanced oxidation or reduction processes, and ball milling (Bolan et al. 2021). Since the goal of the PITT was to identify technologies that would destroy PFAS completely, we focused on destruction methods versus stabilization or removal (e.g., washing or thermal desorption) methods.

Granular activated carbon/anion exchange resins

While ingestion of contaminated food has been identified as the primary PFOA exposure pathway for the general population, drinking water can become the primary exposure pathway in communities with contaminated water (Vestergren and Cousins 2009). To provide protection from long term exposure to PFOS and PFOA in drinking water, the U.S. EPA established drinking water health advisory concentration limits for PFOS and PFOA of 70 ng/L in 2016 (U.S. EPA 2016a, 2016b). To meet these limits, granular activated carbon (GAC) and anion exchange resins (AEX) have been utilized as treatment methods for separating PFAS from liquid streams, often at a fraction of the cost of other separation technologies such as nanofiltration and reverse osmosis (Dickenson and Higgins 2016). GAC and AEX are commonly used to separate PFAS from liquid streams during groundwater site remediation and manufacturing as well as drinking water treatment. GAC removes PFAS compounds from liquid by surface adsorption whereas AEX involves ion exchange onto a positively-charged surface. The surfaces of both materials become saturated over time and no longer able to remove PFAS. GAC can be regenerated but eventually loses its effectiveness and must be disposed. AEX resins used for treatment of PFAS may or may not be amenable to regeneration. Spent GAC and AEX are currently either landfilled or incinerated. The former may lead to PFAS ending up in the landfill leachate.

Aqueous film-forming foams

AFFF are water-based foams that are used to extinguish fires, generally those involving flammable liquids, and are commonly used with aircraft operations. They generally contain a mixture of hydrocarbon and fluorocarbon surfactants, such as fluorotelomers with zwitterionic, cationic,



and anionic properties along with PFOA or PFOS (D'Agostino and Mabury 2014; Place and Field 2012). Fluorinated surfactants' unique characteristics help the foams spread and isolate the fuel from the air, smothering the fire (Moody and Field 2000). The presence of fluorine on the molecules also makes PFAS effective free radical scavengers, which reduce the availability of hydroxyl radicals, the primary chemical component that serves to maintain combustion in conventional fires (Tsang, Burgess, and Babushok 1998).

There is a stockpile of existing AFFF materials, estimated at 9.9 million gallons (United Nations Environment Programme 2004), that now requires disposal to prevent environmental contamination. Most of these older formulations contain C₈ PFAS foams and have been phased out in favor of C₆ PFAS foams (Lim 2019), but still require disposal of the legacy stockpile. Alternate formulations that do not contain PFAS are being developed (Vergun 2019), but disposal of the legacy AFFF is problematic for both the military and civilian stockpiles.

According to a United Nations Environmental Program (UNEP) report United Nations Environment Programme (2004), quantities of AFFF are generally expressed as volume percentages of concentrate. In the foam industry, concentrates are typically referred to as "3%" or "6%" concentrate, depending on the mixture rate with water. For example, if the container of foam concentrate is labelled as 3%, then 97 volumes of water are mixed with 3 volumes of PFAS to make 100 volumes of foam solution for fire application. The high water concentration of AFFF, whether the concentrate or the diluted form, is problematic for multiple types of disposal methods.

On-going ORD research efforts, including projects initiated by the PITT, are underway to identify the temperature and time requirements for effective combustive destruction of AFFF. In addition to these combustion/incineration technologies, less developed, non-thermal approaches were considered by the PITT.

Potential, non-combustion treatment technologies

The PITT considered an array of different treatment technologies to deal with the five PFAS-containing waste streams discussed previously. The PITT specifically focused on promising technologies that were underfunded or understudied and that showed potential to advance with limited time and resources. The team considered various factors in its decision process, including those listed in Figure 1. The selection or

dismissal of technologies was not an indication of the potential of those technologies, as the PITT passed over many promising technologies for a variety of different reasons unrelated to technology potential.

The PITT elected to focus on four emerging technologies, including:

- Mechanicochemical destruction (ball milling)
- Electrochemical oxidation
- Supercritical water oxidation (SCWO)
- Pyrolysis/gasification

Each of these technologies has characteristics that generally align them with specific PFAS-contaminated materials. Research Briefs are available on the PITT's web site, https://www.epa.gov/chemical-research/pfas-innovative-treatment-team-pitt, to explain each of the technologies, their benefits, and areas where more research is needed.

The factors related to treatment selection and process management are shown in Figure 2 for each of the four focus technologies. Selection of an applicable technology must consider how the waste characteristics and processing requirements match up with the capabilities of the technology. Factors related to engineering design considerations are indicated in Figure 2 for each of the four treatment technologies. The size or volume of the system is a function of the treatability goals and effectiveness, and whether the system is meant to be mobile or stationary. Consideration of energy consumption, capital costs, technology complexity, and lifecycle also are required considerations. In some cases, additional pretreatment unit operations such as de-watering or addition of co-mingling agents will either be required or aid in the effectiveness of the technology. Post-treatment unit operations may also be required such as when a byproduct of the process is expected. For example, PFAS breakdown may result in smaller C-number molecules that are volatile, requiring gas treatment to prevent emissions.

- Effectiveness
- Readiness (e.g.: timeline, maturity, capacity)
- Applicability
- Cos
- Specificity (phase and characteristics of the contaminated matrix)
- Safety of outputs (potential for formation of harmful by-products)
- Required development remaining
- Constraints
- Opportunity level for PITT to influence development

Figure 1. Decision factors discussed about all technologies. Four technologies were selected based upon these factors.

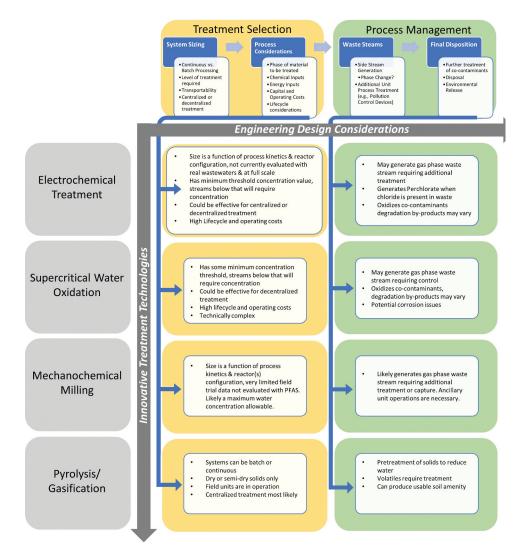


Figure 2. Innovative treatment technology considerations.

Waste-specific characteristics, such as the physical form of the PFAS waste source, are a major determinant of technology suitability. Figure 3 defines the technology selection in terms of the waste factors, handling characteristics, volume, co-contaminants, site locations, and concentrations. The PFAS phase, for example liquid leachate or soil contaminant, is a major factor in technology selection.

Mechanochemical milling

Treatment of solid or semi-solid matrices in high energy ball mills has been proposed as a means of treating PFAS-containing soils and dewatered sludges. Mechanochemical degradation (MCD) has been undertaken on organic-contaminated media (Cagnetta et al. 2016) often with silica or alkaline earth elements to react with fluorine and encourage reactivity. Reactive radicals, electrons, heat, and even plasma in localized areas of the crystalline structures are

created by the impact of stainless steel milling balls in the rotating vessel (Nakayama 2010). Effective destruction (> 99%) of PFAS molecules, including PFOS and PFOA, have been demonstrated in laboratory scale ball mills using pure PFAS compounds and different co-milling reagents (including calcium oxide, potassium hydroxide, lanthanum oxide, sodium persulfate, and sand) (Bolan et al. 2021; Cagnetta et al. 2016, 2017; Lu et al. 2017; Wang et al. 2019; Zhang et al. 2013). Mechanochemical treatments have been effective in soil remediation for chlorinated chemicals (Bolan et al. 2021); however, the treatment of PFAS-containing wastes at a commercially relevant scale (~3 tons per hour) using mechanochemical methods requires additional study. In particular, the effectiveness of MCD at destroying PFAS in soil matrixes (with or without co-milling reagents) remains to be proven.

MCD could be developed in a portable (trailer-based) system or set up on-site for extensive remediation projects, as some commercial companies (e.g., Environmental

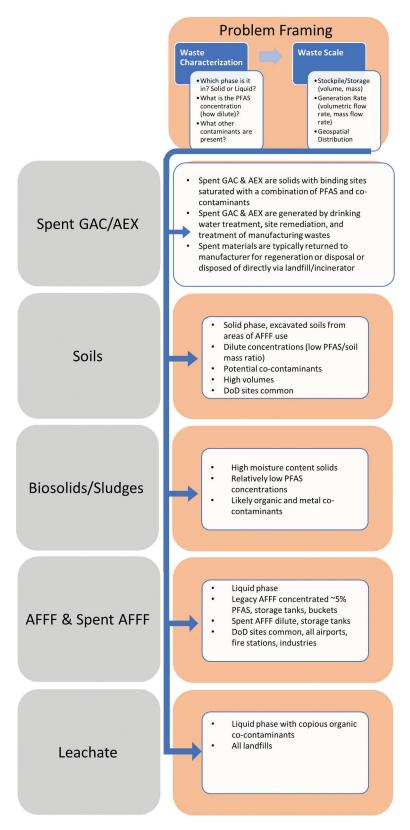


Figure 3. Characteristics of PFAS contaminated matrices.

Decontamination Limited) have done successfully in the past (Bulley 2020). In addition, MCD is unique in that it treats materials in the solid phase without combustion, as compared with most other non-combustion techniques that remove the PFAS from the solid phase into the liquid (e.g., soil washing) or gas (e.g., thermal desorption) phases for treatment. Despite this, it is possible that gaseous PFAS compounds could be released during the MCD treatment, especially in open systems, so a full MCD treatment system must address potential release of volatile PFAS during treatment.

MCD is most amenable to highly contaminated soils, requiring a pre-treatment drying step to remove moisture from the soils, followed by addition of one or more co-milling reagents to react with the PFAS molecules. The resulting soils would be much finer than the initial soils and would lose much of their microstructure. This may affect the fertility of the treated soils. However, mixing treated soils with untreated soils (such as clean soil from off-site) could allow partial recycling of site soils without the potential mechanical and biological issues associated with the milled soils. Another potential issue is the impact of any co-milling reagents on the resulting treated soils. For this reason, non-reactive reagents (such as silicon dioxide or other metal oxides) are preferable to highly reactive (e.g., oxidizing, such as persulfate, or reducing, such as metal hydrides) or pHmodifying (such as acids or hydroxides) reagents. PFAS impacted soils have recently been remediated with and without co-milling reagents in a benchtop ball-mill and showed over 99% destruction of the PFAS with limited byproducts that the authors could analytical resolve (Turner et al. 2021). Further studies are needed to evaluate the gas phase emissions and to better determine any PFAS byproducts.

It is possible that MCD could be used to treat other solid waste streams containing high PFAS concentrations, such as dried GAC. This is especially true with the addition of co-milling reagents to add reactivity and change the mechanical properties of the milled solid. Experiments are required to determine the feasibility of such approaches. More information on MCD can be found in the U.S. EPA's Mechanochemical Degradation Research Brief (Shields and Whitehill 2021).

Electrochemical treatment

Electrochemical oxidation (EO) has been used to oxidize pollutants by means of passing an electrical current through a solution. The electronegativity and electron affinity of fluorine allows the C-F bond to be broken and the fluorine atoms reduced when a high overpotential, > 3 V, is applied to a solution (Niu et al. 2016). The general

destruction mechanism involves a step-wise removal of CF₂ groups and the synthesis of shorter carboxylic acid PFAS until all the carbons have been defluorinated (Le et al. 2019; Niu et al. 2016). PFAS, including PFOA and PFOS, have been destroyed using several different types of electrodes in a search to improve efficiencies and allow for process scale up. Materials such as borondoped diamond (Liao and Farrell 2009; Soriano, Gorri, and Urtiaga 2017), titanium suboxides (Le et al. 2019; Wang et al. 2020), and tin and lead oxides (Niu et al. 2016) have been used to successfully degrade PFAS in aqueous solutions. The process has been demonstrated on PFAS at the bench and pilot scale (Liang et al. 2018; Nzeribe et al. 2019). The technology has advantages of being able to operate at ambient temperature and pressure. EO is diffusion limited and the remediation tends to slow as the concentration of PFAS decreases, so innovative electrode and reactor designs are needed to allow for scale-up. Other issues involve the buildup of minerals on the anode, possible formation of perchlorate and other inorganic by-products, and the generation of volatile by-products which need to be treated. Investigations into the use of an electric field to transport and destroy PFAS in soils have been started and show promise (Skinn 2019) as have tests on landfill leachate (Pierpaoli et al. 2021) and AFFF contaminated groundwater (Schaefer et al. 2019). Further information on this relatively low-energy technology is available in the U.S. EPA's Electrochemical Oxidation Research Brief (Krause, Magnuson, and Crone 2021).

Supercritical Water Oxidation (SCWO)

Above 374°C and 22.11 MPa, water reaches its supercritical state in which it behaves as a solvent for organics and enhances chemical oxidation reactions in the presence of an oxidizing agent such as O2, air, or hydrogen peroxide. SCWO has been demonstrated to treat halogenated waste materials including polychlorinated biphenyls (Abeln et al., 2001; Kim et al. 2010b). SCWO is able to quickly treat not only water based waste streams, but also sludges (Svanström et al. 2004) and slurries (Kim et al. 2010a); providing a possible way to treat most any pumpable matrix. Although SCWO requires initial energy input to reach the required temperature and pressure, the resulting reactions often produce a significant amount of heat themselves, which can be harvested to keep driving the process. The concentration of oxidizable organic molecules in the material being processed must be controlled and moderated, either by diluting the influent stream or adding additional fuels (such as alcohols). During the oxidation process, SCWO produces many acidic species, such as



sulfuric acid (from sulfur-containing species) and hydrochloric acid (from chlorine-containing species). This results in a drop in pH during the process, which can cause significant corrosion of the reactor if not addressed. This can be addressed to some extent by adding alkalinity to the influent solution. The increased alkalinity of the modified influent solution may aid in the destruction of PFAS, as alkaline solutions show greater destruction and mineralization of PFOS at subcritical conditions than acidic or neutral solutions (Wu et al. 2019). An additional challenge of SCWO is the precipitation of salts. The three major product streams from a SCWO reactor are the effluent, any evolved gases, and the salts that precipitate out from the reaction. A thorough evaluation of PFAS destruction using SCWO must address all of these product streams. Secondary air treatment technology might be necessary if gaseous PFAS compounds are evolved as a result of the SCWO process. There are different manifestations of SCWO based on different process designs. The PITT explored four different manifestations of SCWO technology to evaluate the potential for PFAS destruction.

Technical challenges being addressed when creating commercial products (374water 2020; Battelle Memorial Institute 2019; General Atomics, industrial Supercritical Water Oxidation (iSCWO) 2021; Jama et al. 2020) for the widespread implementation of SCWO include the buildup of corrosive gases during the oxidation reaction, the precipitation of salts, and the high energy requirements. More information is available in the U.S. EPA's Research Brief on SCWO (Sahle-Damesessie and Krause 2021).

Pyrolysis/gasification

Pyrolysis decomposes solid or semi-solid materials at temperatures typically in the 300°C to 1000°C range in an oxygen-free environment. Gasification is similar to pyrolysis but operates at temperatures typically in the 800°C to 1650°C range with substoichiometric quantities of oxygen in a partial combustion process to provide additional energy to the process (Fytili and Zabaniotou 2008; Patel et al. 2020; Winchell et al. 2020). Pyrolysis can form a useful char (Boni et al. 2021) product whereas gasification typically forms low carbon ash. Both processes can generate a hydrogen-rich synthetic gas (syngas) depending on operating conditions. The high temperatures and residence times achieved by the combination of pyrolysis or gasification, followed directly by combustion of the hydrogen-rich syngas stream in a thermal oxidizer could potentially destroy PFAS by breaking apart the chemicals into inert or less recalcitrant constituents, although this remains a subject further research. Compared to traditional

incineration, pyrolysis and gasification require much lower air flows than incineration, which reduces the size and capital expense of air pollution control equipment. More information is available in the U.S. EPA's Research Brief on Pyrolysis and Gasification of Biosolids (Acheson et al. 2021).

Waste source and treatment technology applicability

Some of the basic factors and characteristics involved in matching PFAS waste sources with these four innovative technologies are illustrated in Figure 2 for the technology focus and Figure 3 for the waste focus. These illustrations are meant to serve as a general guide to technology selection, management of the processes, and issues to consider PFAS waste characteristics.

A crosswalk between the technology factors for these four treatment methods and the PFAS waste characteristics, coupled with a literature review on the level of technology development, is presented in Figure 4. These factors are synthesized into an assessment of Technology Readiness Level (TRL, see Figure 4 Key in Figure 5) by the PITT to understand both the waste type applicability and state of development of each technology. These assessments were made by the PITT based on available information and the team's best judgement and should be considered a starting point for discussion and technology development prioritization.

Spent GAC/AEX are solids that may be applicable to treatment in MCD or pyrolysis/gasification systems but are not applicable to liquid-based waste treatment technologies such as SCWO or EO. To our knowledge, no PFAS-laden GAC/AEX tests with MCD and pyrolysis/gasification have been undertaken. The crushing and grinding of the MCD systems seem an appropriate test for the solid GAC media. The elevated temperatures in pyrolysis/gasification systems may be an issue for the polymer-based AEX resins, causing melting and fouling of internal systems.

Like GAC/AEX, PFAS-contaminated soils could be treated in an MCD or pyrolysis/gasification unit. Both systems are ideally suited for treatment of solid materials. MCD has been used to treat soils contaminated with polychlorinated biphenyls, achieving over 99% mass reduction of soil contamination (Bulley 2020). We could not find literature references to pyrolysis/gasification treatment of PFAS-laden soils. However, the material handling characteristics and treatment operations are similar to those used in thermal decontamination of PFAS-contaminated soils (Bolan et al. 2021; Crownover et al. 2019; Sörengård et al. 2020). Therefore, although the

	Electrochemical Oxidation	SCWO	Mechanochemical degradation	Pyrolysis
Spent GAC / AEX	TRL 4 ^a - AEX still bottom	N/A ^j	TRL 2 ^{i, j}	TRL 1 ^j
Soils	TRL 1 ^b	N/A ^j	TRL 5/6 ⁱ	TRL 1 ^j
Biosolids / Sludges	N/A j	TRL 6 ^e	TRL 1 ^j	TRL 7 ^k
Spent and unused AFFF	TRL 4/6 ^{a, c}	TRL 7 ^{e-h}	TRL 3/4 ⁱ	N/A ^j
Landfill Leachate	TRL 4 ^d	TRL 4 ^g	N/A ^j	N/A ^j

- a. AECOM (Liang 2018); b. Faraday (Skinn 2019); c. (Schaefe, 2019); d. (Pierpaoli 2021);
- e. (374Water 2020); f. (General Atomics 2021); g. Aquarden (Jama 2020); h. (Battelle 2019);
- i. EDL (Bolan 2021); j. PITT assessment; k. (BioForceTech 2019)

Figure 4. Crosswalk between PFAS waste and innovative Technology Readiness Level (TRL). Colors correspond to TRL categories described in Figure 5. No color indicates the non-applicability (N/A) of the technology to the waste stream.

Phase	TRL	Description		
- U	1	Basic Principles observed		
Research	2	Technology concept formulated		
	3	Experimental proof of concept		
nent	4	Technology validated in lab		
Development	5	Technology validated in relevant environment		
	6	Technology demonstrated in relevant environment		
ent	7	System prototype demonstration in operational environment		
Deployment	8	System complete and qualified		
Dep	9	Actual system proven in operational environment		

Figure 5. Key for Figure 4 technology readiness levels. Note that technologies were rated for PFAS destruction and may not capture their full capacity or overall readiness-level for other treatment purposes. Source: https://www.twi-global.com/techni cal-knowledge/faqs/technology-readiness-levels.

release and treatment of volatiles requires additional study, the treatment of PFAS-contaminated soils using pyrolysis or gasification is technically feasible.

Biosolids/sludges are likely candidates for treatment in MCD, pyrolysis/gasification, and SCWO systems. Sufficiently dewatered sludge, or wet sludge dried by addition of co-milling agents, would be applicable to treatment in an MCD system. No such tests with PFAS have been identified in the literature. Preliminary studies on sludge treatment with SCWO have shown strong reductions in PFOS and PFOA levels in the processed effluent (374water 2020). PFAS testing on a biosolids pyrolysis system (Bioforcetech Corporation 2019) was repeated in a test commissioned by the PITT confirming high levels of degradation of the target PFAS compounds analyzed in the feed. Results on the latter are expected to be published in 2021.

A common source of stockpiled, legacy AFFF concentrate has been designated for upcoming SCWO, EO, and laboratory-scale MCD testing by the PITT with four SCWO providers. This legacy AFFF is comprised of 8-carbon PFAS, acids, and zwitterions as well as nonfluorinated surfactants and stabilizing chemicals (Material Safety Data Sheet FC-203CF 1996). Preliminary results from these tests show up to 99% degradation of the initial ("target") PFAS compounds in the feed. However, in all cases, the potential non-target byproducts have yet to be analyzed.

PITT efforts

A variety of mechanisms have been put into place to test AFFF destruction using SCWO with four commercial companies. Where feasible, a common AFFF source has been used for these tests. These studies have achieved preliminary results; complete studies on emissions and byproducts are underway. Hydrothermal oxidation, the lower pressure, lower temperature variant of SCWO, is undergoing both laboratory scale tests at EPA and pilotscale commercial tests (as a SCWO pretreatment step). EO of dilute AFFF is undergoing testing in a laboratory scale reactor, accompanied by collection and analysis of the off-gases for volatile fluorinated organics. Laboratory scale studies of MCD of AFFF-laden soil are being conducted at EPA and a commercial laboratory. Scale-up plans and emission sampling tests have been proposed to extend this effort. Sludge gasification studies have been conducted at a field unit. The destruction of inlet PFAS is being assessed as well as the potential for fluorinated compound emissions. Preliminary results from all four technology/waste tests are expected to be available by 2022.



Summary

Four non-combustion technologies were highlighted for their potential to treat PFAS-laden waste streams: electrochemical oxidation, mechanochemical degradation, supercritical water oxidation, and pyrolysis/gasification. Considerations regarding technology factors and waste characteristics were cross-walked to aid in selection of treatment options. Finally, the technology applicability and readiness level were presented based upon existing, published information. Refinement of technology selection will be dependent on additional data; currently, only extremely limited testing results are available. Programs put into place by the PITT to examine these technologies are expected to yield published results by the end of 2022.

Acknowledgment

Funding for this work was provided by the U.S. EPA Office of Research and Development. The authors claim no competing interests. The views expressed in this paper are those of the authors and do not necessarily represent the views or policies of the U.S. Environmental Protection agency.

Disclosure statement

No potential conflict of interest was reported by the author(s).

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